

# SAFETY AND RISK MANAGEMENT IN NUCLEAR-BASED HYDROGEN PRODUCTION WITH THERMAL WATER SPLITTING

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## ABSTRACT

The challenges and approaches of the safety and risk management for the hydrogen production with nuclear-based thermochemical water splitting have been far from sufficiently reported, as the thermochemical technology is still at a fledgling stage and the linkage of a nuclear reactor with a hydrogen production plant is unprecedented. This paper focuses on the safety issues arising from the interactions between the nuclear heat source and thermochemical hydrogen production cycle, as well, between the proximate individual processes in the cycle. As steam is utilized in most thermochemical cycles for the water splitting reaction, and heat must be transferred from the nuclear source to hydrogen production plant, this paper particularly analyzes and quantifies the heat hazard for the scenarios of start-up and shutdown of the hydrogen production plant. Potential safety impacts on the nuclear reactor are discussed. It is concluded that one of the main challenges of safety and risk management is efficient rejection of heat in a shutdown accident. Several options for the measures to be taken are suggested. Copper-chlorine and sulphur-iodine thermochemical cycles are taken as two representative examples for the hazard analysis. It is expected that these newly reported challenges and approaches could help build the future safety and risk management codes and standards for the infrastructure of the thermochemical hydrogen production.

## 1. INTRODUCTION

Nowadays, hydrogen is a necessity for the production of fertilizer and upgrading of heavy oils, with the demand increasing and growing rapidly. Hydrogen is a promising alternative fuel for our future vehicles and home heat supply. Especially, if hydrogen is produced from clean energy, the usage of hydrogen would reduce pollutant and greenhouse gas emissions that are detrimental to our atmosphere and climate [1]. Currently, the hydrogen production is mainly produced from fossil fuels, e.g., steam methane reforming and coal gasification [2]. To achieve a clean hydrogen production, researchers have been developing new methods in the past decades. Among the methods, high temperature electrolysis (HTE) and thermochemical hydrogen production cycles using clean energy to split water thermally through intermediate chemical compounds and reactions have attracted more and more interest [3-8]. Nuclear energy is considered as a major thermal energy source to meet the heat requirement of high temperature electrolysis and thermochemical cycles.

There are still many challenges hindering the fast industrialization of high temperature electrolysis and thermochemical cycles, although these methods are viewed as promising options for the future clean fuel supply, they are unprecedented and no sufficient industrial operation experience has been acquired, particularly from the perspective of operation safety when a hydrogen production plant is linked with a nuclear reactor. Traditional nuclear safety research has been focusing on the safety of nuclear reactor core and its thermal hydraulic system, and the chemical plant safety has examined the probability of fires, blasts, leaks, and spills [10]. However, the likelihood of obtaining a permit to construct such a nuclear and hydrogen combination without proposing measures to be taken for avoiding risks and accidents is questionable, because the linkage may result in complex mutual interactions between the hydrogen production and nuclear power plants. In other words, the safety assurance must consider the mutual interactions and not be only limited to the hydrogen production plant.

A number of investigators have speculated about some disastrous situations when a fire or blast of hydrogen takes place. The worst scenario could be a severe damage or detonation of nuclear reactor core caused by the blast of hydrogen plant [9-11]. The probability of detonation was quantified, and a minimum separation distance between nuclear reactor and hydrogen production units was suggested to avoid the detonation of nuclear core. To minimize the risk, some measures were proposed [10, 11], including putting a 100 kg on-site limit for Hydrogen storage, quickly piping hydrogen out as produced, location of the nuclear plant control room outside of the dispersion zone for chemical release. Detrimental impacts of the chemicals in a hydrogen production plant on the nuclear reactor were examined qualitatively and some risk-mitigating design modifications were proposed to minimize the probability of nuclear core damage. The modifications include constructing an earthen barrier between the nuclear and chemical facilities, constructing the nuclear facility primarily underground, constructing blast panels near the chemical facility to dampen overpressure events, constructing the chemical facility primarily underground and moving the nuclear plant control room offsite.

The past research of the safety issues has put significant forward-looking contributions to the nuclear-based hydrogen production. However, there are still too many other safety issues yet to be examined. As heat must be transferred to thermochemical cycles and high temperature electrolysis, this paper will quantitatively examine the hazards caused by the interactive heat exchange between the nuclear and hydrogen production plants from the perspectives of heating fluid fail and heat rejection, through adopting a top-down approach to identify the system boundary. A big difference between thermochemical water splitting and steam methane reforming (SMR) for hydrogen production is that a thermochemical cycle usually consists of more chemical processes than SMR. When these processes are integrated together to form a closed loop, their interactions may also complicate the safety issues. This paper will also examine the influences of these interactions on the safety.

## 2. HEAT HAZARDS RELATED TO NUCLEAR REACTOR – H<sub>2</sub> PLANT HEAT EXCHANGE

### 2.1 Material and Energy Flows across the Boundary between Nuclear Reactor and H<sub>2</sub> Plant

Before looking into the details of nuclear reactor and hydrogen production plant, a top-down approach can be used to study the linkage of nuclear reactor and hydrogen production plant so that the material and energy flows across their boundary and the mutual impacts can be expressed in a simplified chart. Figure 1 shows the material and energy flows.

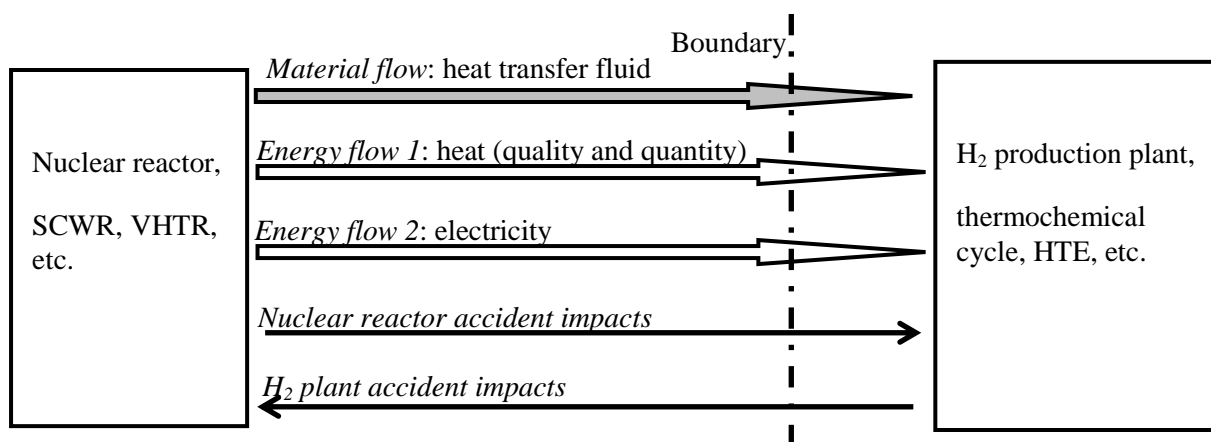
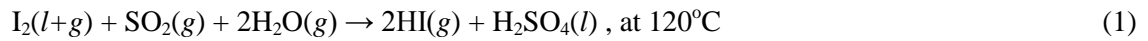


Figure 1. Material and energy flows across the boundary between a nuclear reactor and H<sub>2</sub> plant

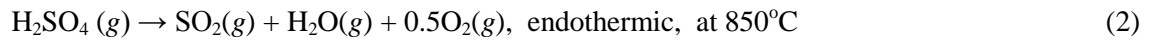
Figure 1 shows that there are one material and two energy flows across the boundary. In this paper, the study of the mutual interactions starts from examining the flows. The energy flows include heat and electricity. Since a hybrid thermochemical cycle consists of both electrolytic and fully thermal processes, the electricity usage here means the electricity to be utilized to support the electrolytic reactions required by hybrid thermochemical cycles and high temperature electrolysis, rather than drive the pumps and control units that are also existing in other hydrogen production methods. From this point of view, if a fully thermal water splitting cycle is used, no electricity passes across the boundary.

A fully thermal water splitting cycle is sulfur-iodine (S-I) cycle [5, 6] consisting of the following reactions summing to the water splitting:

Step 1: Hydrolysis step (exothermic)



Step 2: Oxygen production step (endothermic)

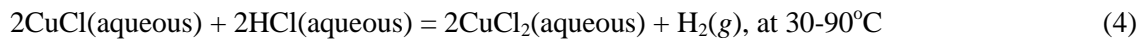


Step 3: Hydrogen production step (endothermic)

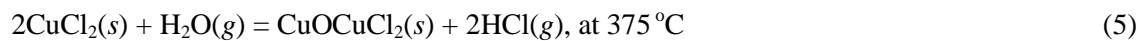


A hybrid thermochemical water splitting cycle is copper-chlorine (Cu-Cl) cycle [7, 8] involving three reactions:

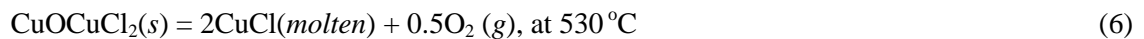
Step I: Electrolytic step for hydrogen production



Step II: Hydrolysis step (endothermic)



Step III: Oxygen production step



Common step for most thermochemical cycles:



About two hundred thermochemical cycles have been reported [4-8]. The reason to take S-I and Cu-Cl cycles to represent fully and hybrid thermochemical cycles is that the scale-up of S-I cycle has been demonstrated by the Japan Atomic Energy Agency (JAEA) [5, 6], and the scale-up of the Cu-Cl cycle from proof-of-principle to a larger engineering scale is under construction at the University of Ontario Institute of Technology in collaboration with its partners that include Atomic Energy of Canada Limited (AECL) and Ontario government [7, 8].

Equation (4) indicates the electricity requirement in the hybrid Cu-Cl cycle. According to past studies, the electricity occupies less than 20% of the total energy requirement [12]. Also, since outputting electricity from a nuclear power plant is a mature technology and the electricity could be replaced by another source from a power grid in case the nuclear electricity supply fails, this paper will not focus on the safety and risk issues related to the electricity.

## 2.2 Intermediate Working Fluids for a Safe and Reliable Heat Transfer

In theory, nuclear reactor coolant could provide the required heat to thermochemical cycles for hydrogen production. However, the coolant is not recommended as the working fluid passing through the thermochemical cycles to transfer heat to the chemical reactions, because of potential hazards of very high pressure and radioactivity [10, 13]. Therefore, an intermediate heat exchanger (HX) is needed and it will handle nuclear reactant on one side and another heat transfer fluid on the other side. This is conceptually illustrated in figure 2. To assure the safety, the selection of an appropriate heat transfer fluid is very important.

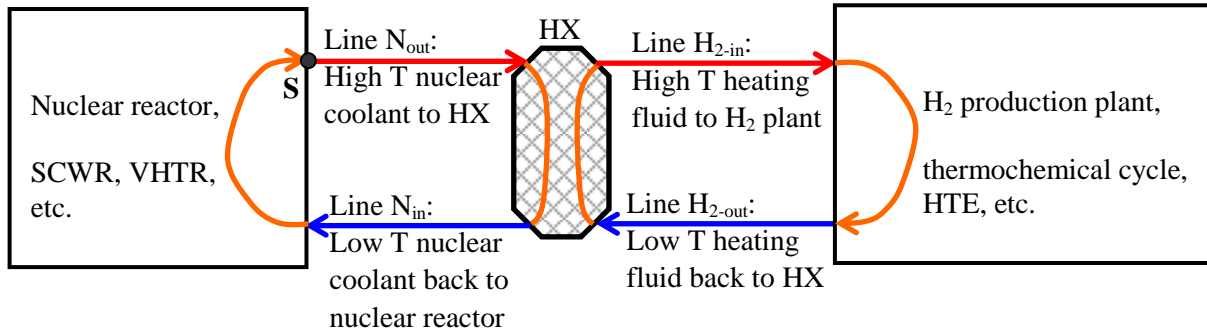


Figure 2. Intermediate heat exchanger (HX) isolating nuclear reactor and hydrogen production plant

Past safety studies of heat transfer fluids have considered the anticorrosion, burst pressure tolerance, and radioactivity contamination. In addition to these considerations, this paper puts more speculations on the safety issues related to the selection of working fluids from the perspectives of liquid clogging resulting from phase change, steam explosion due to the external water leak, and choking and dispersion hazards when leaking into air. Table 1 summarizes these considerations.

Table 1. Safety issues related to heating fluids

Safety considerations		Heating fluids				
		molten salts	helium	steam	supercritical H <sub>2</sub> O	supercritical CO <sub>2</sub>
1	Pressure blast due to high pressure requirement.	No	Yes	Yes	Yes	Yes
2	Corrosion / abrasion to pipe materials.	Yes	No	Possible	Possible	Possible
3	Clogging resulting from phase change.	Yes	No	Yes	Moderate	No
4	Steam explosion if water leaks into the transport pipeline.	Yes	No	No	No	No
5	Choking and dispersion hazards if leaking into air.	No	No	Moderate	Moderate	Yes
6	Likelihood of leak.	low	high	low	low	low
7	Nuclear activation.	No	Possible	No	No	No

In the first safety consideration of pressure blast due to high pressure requirement, molten salts do not have this issue because the volumetric heat capacity of molten salts low dependency on pressure and is usually several orders larger in magnitude than that of gases. In addition, molten salts have low viscosity and can flow with minimal pumping power compared with gases [10]. In comparison, to possess an equivalent heat capacity, gases must be pressurized. This may result in pressure blast if

there is a failure in the heat exchanger and heating fluid transport pipes, especially if the pipes are constructed over a long distance. From equations (2) and (6), it can be found that the pipe burst pressure should function well at a temperature of higher than 530°C for Cu-Cl cycle, and 850°C for S-I cycle.

In the second safety consideration, i.e., the corrosion to pipe materials, helium is expected to perform best among the fluids in Table 1, because it is a noble gas and hence inert to most materials. In comparison, the corrosion attack intensity of steam, supercritical water, and supercritical CO<sub>2</sub> depend on temperature and pipe materials. As to the molten salts, the research has been performed in the past decades on the chemical corrosion and abrasion phenomena [14, 15], there are still many unknowns today and the molten salt chemistry plays a major role in the viability demonstration of molten salt nuclear reactor [16, 17]. So it is concluded that the intermediate heat exchanger may have the same viability challenge if using molten salt as the second heat transfer fluid.

As for the third safety consideration of pipe clogging resulting from phase change, as shown in figure 2, the working fluid temperature of Line H<sub>2-out</sub> is lower than that of Line H<sub>2-in</sub>. The temperature drop may cause the formation of liquid coming out of gas or solid coming out of liquid, due to the condensation or solidification after the heating fluid exits the hydrogen production loop. As indicated by equations (1) – (6), the temperature in Cu-Cl cycle drops from 530 to 375°C, and 850 to 450 °C in S-I cycle. If the steam reactant of equation (7) is generated at near-atmospheric pressures, the exiting temperature would be much lower. The phase change may also be caused by low temperature local spots at the wall of heating fluid transport pipeline, as well as valves and expansion joints, which have been observed by the experiments in the Clean Energy Research Laboratory in University of Ontario Institute of Technology. This may significantly increase the possibility of condensation or solidification of the working fluid. To avoid the risk, the working fluid should be non-condensable, or the phase change is negligible. Table 2 shows the comparison of the minimum temperatures at the outlet (Line H<sub>2-out</sub> of figure 2) of thermochemical cycles and the critical, liquefaction, and solidification points of different fluids.

Table 2. Minimum temperatures at the outlet (Line H<sub>2-out</sub> of figure 2) of thermochemical cycles vs. critical, liquefaction, and solidification points of different fluids.

Minimum outlet temperatures of thermochemical cycles	Properties		Heating fluids			
			molten salts <sup>(b)</sup>	helium	H <sub>2</sub> O	CO <sub>2</sub>
450°C for S-I cycle in equation (3)	Critical point	°C	N/A	-267.96	373.85	31.1
		MPa	N/A	0.227	22.06	7.38
375°C for Cu-Cl cycle in equation (5)	Liquefaction / boiling point, °C		1430	-268.93	99.98	-78
100-130°C for both cycles <sup>(a)</sup> in equation (7)	Solidification / melting point, °C		400	-272.20	N/A	N/A
(a) If steam is generated at near-atmospheric pressures.						
(b) The molten salts are chlorides and/or fluorides [[14-17].						
(c) N/A means “not applicable” because the properties are unknown or meaningless to the applications of interest here.						

From Table 2, it is observed that helium is again expected to perform best among the fluids to avoid a clogging caused by phase change, because its critical point is too low (5.19K) to be considered condensable in engineering practice. CO<sub>2</sub> could be regarded as the second best working fluid because its critical temperature is near ambient and much lower than water. Steam is not recommended as a working fluid because its critical temperature is about 374°C and hence easy to condensate at a lower temperature, except supercritical water is utilized as the working fluid. However, this needs a minimum pipeline pressure of 22MPa. At this pressure, not only the pipe material cost will go up, but the risk of pressure burst is also increased. Using a molten salt is a good option for the avoidance of

pressure blast, but the molten salts under current investigations may be subject to solidification due to the high solidification point of 400°C. Therefore, to find a molten salt with low solidification point is a part of the future research.

The fourth safety consideration listed in Table 1 is steam explosion of the heating fluid transport pipeline. It has been reported that serious steam explosion may take place when liquid water is poured onto the surface of a liquid metal or salt [18-20]. This paper speculates on this issue because the pipeline may be constructed over a long distance and the leak of liquid water into the pipeline due to seismic or corrosive causes may occur. If the working fluid is a molten salt, then the water leak may result in serious steam explosion. Other working fluids such as helium, supercritical water, and CO<sub>2</sub> do not have this safety issue.

The fifth safety consideration in Table 1 is the choking and dispersion hazards if the working fluid flowing in the pipeline leak into ground surface and air. Except molten salt, other fluids will enter air. If the leak is not serious, the leaked fluid may be diluted soon. However, if the leak is significant, the working fluid may not have sufficient time to be diluted. In this situation, CO<sub>2</sub> may cause choking hazard due to its larger density than air. In comparison, helium causes the slightest choking hazard because it is much lighter than air and rises very fast departing the ground surface. The choking hazard of steam is hard to decide, because it depends on many factors such as the leak rate and wind speed.

The last safety consideration listed in Table 1 is the possibility of nuclear activation. It was reported [10] that using helium as either a coolant or a heat transfer fluid may raise the possibility of tritium generation through neutron activation of He-3. This is a weakness of utilizing helium as the heating fluid.

### **3. HEAT REJECTION FOR SAFE START-UP AND SHUTDOWN OPERATIONS**

Past studies of the linkage of nuclear power and thermochemical hydrogen production plants have focused on steady state operation, i.e., the hydrogen production and heat transfer rates do not change with time. The transient behaviour and the related dynamic safety issues arising from the linkage have been rarely investigated. In the start-up of a thermochemical hydrogen production plant, the hydrogen production rate cannot jump from zero to its designed production rate abruptly. Thus, the rate of heat transfer from the nuclear reactor to hydrogen production plant must match the transient heat requirement of the hydrogen production in the start-up operation. Similarly, when the hydrogen production plant encounters an accident or maintenance shutdown, the heat supplied from the nuclear reactor to the hydrogen production plant must be modulated to match the shutdown requirement. As indicated by equations (1)-(7), a thermochemical cycle consists of several integrated chemical steps, the failure of any step may incur the shutdown of other up and downstream steps. So it is beneficial to speculate the emergency shutdown scenario so that the safety of both nuclear and hydrogen production plants can be assured.

Since the heat supply capacity of nuclear reactor is designed for a full capacity hydrogen production, surplus heat must have somewhere to go because no surplus heat is allowed in the nuclear reactor core, otherwise the reactor core may collapse due to the high temperature caused by the surplus heat. Obviously, it is not accepted to release working fluid to ambient air so as to stop the heat supply to the hydrogen plant, because this may cause a quick accumulation of surplus heat in the nuclear core and result in meltdown of nuclear reactor core. This paper proposes five measures that could be potentially taken for the rejection of the surplus heat, as indicated by R1 – R5 in Figure 3.

The first measure (R1, the route connecting points A, B and C) is to conduct the surplus heat to turbine for power generation, and the power could be uploaded to power grid. The second measure is to reject the heat to the heat sink designed for the nuclear coolant, e.g., river or lake. This would need an extra design capacity for the heat sink equipment. For example, the processing capacity of the condenser of the power plant should be increased to accommodate the heat rejection rate in case of an emergency

shutdown of hydrogen production plant. Both heat rejection measures R1 and R2 have a common weakness: the working fluid in the transport pipeline (the route connecting points F and G) may subject to a condensation or solidification due to insufficient heat. As a consequence, clogging may occur, which is a safety concern that has been discussed previously.

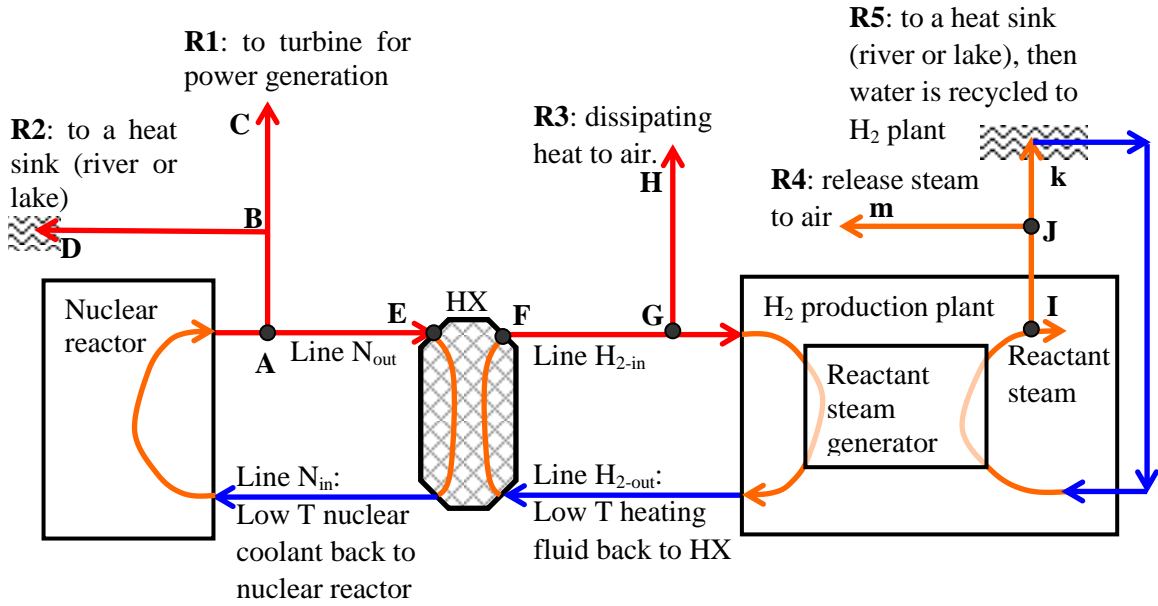


Figure 3. Five options (R1-R5) for the rejection of the surplus heat for a safe start-up and shutdown

To avoid the clogging safety issue, the surplus heat could be rejected on the other side of the intermediate heat exchanger. For example, a bypass heat dissipation structure could be set up on the pipeline, then the heat could be dissipated to air, as indicated by measure R3 (the route connecting points G and H). Another option is to release the generated steam reactant to air, which is indicated by measure R4 (the route connecting points I, J and M), as steam reactant must be generated in most thermochemical cycles. This option has a significant downside: the high purity water is lost to the environment. To overcome this weakness, the steam could be conducted to a condenser rejecting heat to a heat sink such as a river or lake, then the water could be recycled back to the hydrogen production plant to continuously absorb heat and then reject surplus heat. This option is indicated by measure R5 (the route connecting points I, J and K).

To further assess the five measures of figure 3, particular R3, R4 and R5, this paper quantifies the maximum surplus heat in the start-up and shutdown operations, which is listed in Table 3. To calculate the heat quantity, an overall hydrogen production efficiency of 50% is assumed on the basis of reported data for Cu-Cl and S-I cycles [7]. For convenience of conceptually applying Table 3 to other thermochemical cycles, the heat transfer rate for an overall efficiency of 100% is also listed. The overall efficiency  $\eta$  is defined as follows:

$$\eta = \frac{HHV}{\Delta H_{NetInput} + \Delta E_{Electrolysis}} \times 100\% \quad (8)$$

where HHV is the higher heating value of hydrogen (286 kJ/mol),  $\Delta E_{Electrolysis}$  is the electrical energy required for a hybrid cycle with the presence of electrolysis.  $\Delta H_{NetInput}$  is the net heat input to the cycle and its value is based on point S on Line N<sub>out</sub> at the outlet of nuclear reactor, as shown in figure 2.

Equation (8) can apply to fully and hybrid thermochemical cycles as well as high temperature electrolysis. If  $\Delta E_{electrolysis}$  is zero, it means a thermochemical process only utilizing thermal energy to

split water. If neither is zero, it is a hybrid process. As previously discussed, S-I cycle of interest in this paper is a fully thermochemical cycle utilizing thermal energy only, and Cu-Cl cycle is a hybrid cycle. For a simplification of the heat hazard assessment,  $\Delta E_{Electrolysis}$  is appointed a value of zero in this paper for the Cu-Cl cycle, because the electricity consumption is not significant compared with the thermal energy requirement.

Table 3. Heat quantity to be safely rejected in start-up and shutdown operations

H <sub>2</sub> production rate, ton/day	Heat quantity, MW <sub>th</sub>		Equivalent Electricity, MW <sub>e</sub>	Steam (100°C) release rate <sup>(c)</sup> , m <sup>3</sup> /s	Hot air (80°C) generation rate, m <sup>3</sup> /s
	$\eta = 100\%$ <sup>(a)</sup>	$\eta = 50\%$	Power generation efficiency <sup>(b)</sup> : 45%		
0.001	0.00165	0.00329	0.00148	0.00121	0.017
1	1.65	3.29	1.48	1.21	17
10	16.5	32.9	14.8	12.1	170
100	165	329	148	121	1,704
200	329	658	296	243	3,408
500	823	1,646	741	607	8,520
800	1,316	2,633	1,185	971	13,632
1000	1,646	3,291	1,481	1,214	17,039

(a)  $\eta$  represents the overall H<sub>2</sub> production efficiency defined by equation (8).  
(b) The power generation efficiency is based on Generation IV nuclear reactors [21].  
(c) The steam release rate means the steam generation rate with the surplus heat.

It can be found from Table 3 that the heat rejection rate is quite high for an industrial hydrogen production scale. For example, the heat to be rejected for a hydrogen production rate of 500 ton/day, i.e., a rate equivalent to an industrial scale steam methane reforming, the surplus heat is equivalent to that of a Generation IV nuclear reactor. If the surplus heat is conducted to turbine for power generation, the power grid should have a margin for the upload. Table 3 also suggests that measure R3, i.e., dissipating surplus heat to air, is not feasible because hot air (80°C) is generated very fast in a surprisingly large amount (8,520 m<sup>3</sup>/s for a hydrogen production rate of 500 ton/day), which is a significant heat hazard. To release steam in measure 4 is not feasible either, because the steam (100°C) is also generated in a surprisingly large rate (607 m<sup>3</sup>/s for a hydrogen production rate of 500 ton/day), although it is feasible for a laboratory hydrogen production scale due to the small steam quantity to be rejected (0.00121 m<sup>3</sup>/s). The Clean Energy Research Laboratory in University of Ontario Institute of Technology adopts this direct steam release for a laboratory scale heat rejection. In conclusion, using a condenser to reject the surplus heat to a river or lake is potentially the safest option for the start-up and shutdown operations of the hydrogen production plant.

#### 4. CONCLUSIONS

This paper reviews and examines the challenges and approaches of the safety and risk management for the hydrogen production with nuclear-based thermochemical water splitting, and potential safety impacts arising from the hydrogen production on the nuclear reactor are discussed. The paper focuses on the safety issues arising from the interactions between the nuclear heat source and thermochemical hydrogen production cycle, particularly the safety issues related to the heating fluids and the heat



hazards arising from the transient operations such as start-up and shutdown of the hydrogen production plant. Copper-chlorine and sulphur-iodine thermochemical cycles are taken as two representative examples for the hazard analysis. The safety performance of several typical heating fluids is evaluated from the perspectives of pressure blast, corrosion, abrasion, clogging, steam explosion, choking and dispersion hazards, likelihood of leak, and nuclear activation. One of the main challenges of safety and risk management is efficient rejection of heat in an emergency shutdown. Several measures are proposed and compared for a safe rejection of heat. It is expected that these newly discussed safety issues and measures could improve the future safety and risk management codes and standards for the infrastructure of the nuclear-based thermochemical hydrogen production.

## 5. ACKNOWLEDGEMENTS

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